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EVALUATION OF RADIONUCLIDE ACCUMULATION IN SOIL DUE TO LONG-TERM IRRIGATION

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ABSTRACT

Radionuclide accumulation in soil due to long-term irrigation is an important part of the model for predicting radiation dose in a long period of time. The model usually assumes an equilibrium condition in soil with a constant irrigation rate, so that radionuclide concentration in soil does not change with time and can be analytically solved. This method is currently being used for the dose assessment in the Yucca Mountain project, which requires evaluating radiation dose for a period of 10,000 years. There are several issues associated with the method: (1) time required for the equilibrium condition, (2) validity of constant irrigation rate, (3) agricultural land use for a long period of time, and (4) variation of a radionuclide concentration in water. These issues are evaluated using a numerical method with a simple model built in the GoldSim software. Some key radionuclides, Tc-99, Np-237, Pu-239, and Am-241 are selected as representative radionuclides. The results indicate that the equilibrium model is acceptable except for a radionuclide that requires long time to accumulate in soil and that its concentration in water changes dramatically with time (i.e. a sharp peak). Then the calculated dose for that radionuclide could be overestimated using the current equilibrium method.

INTRODUCTION

The surface soil model in the environmental dose assessment is to evaluate radionuclide accumulation in, and removal from, the upper layer of the soil (down to the tilling depth) where all plant roots are assumed to be located. Surface soil is contaminated as a result of using contaminated groundwater for irrigation. Radionuclide concentrations in the soil will build up at a rate determined by the physical and chemical properties of the soil and the radionuclides. On land irrigated for a long time, radionuclide concentrations depend on the rate of accumulation and removal, and they will reach saturation (equilibrium) concentrations when the rates of addition and removal are equal. Long-lived radionuclides that bind readily to soil particles will not reach equilibrium concentrations for thousands of years, whereas relatively short-lived or mobile radionuclides will approach equilibrium concentrations after only a few years.

Radionuclides can be removed from the surface soil by leaching into the deep soil, surface soil erosion, crop harvest removal, and radioactive decay. Although crop harvesting may be an important mechanism for radionuclide removal from agricultural lands, this mechanism is not considered in the model because it is considered to compensate for the reintroduction of radionuclides into the soil when contaminated cow manure is used as fertilizer.

Because irrigation rates differ among crop types, radionuclide concentrations in the surface soil depend on the specific use of cultivated land. However, crop rotation over a longer period of time would average out the radionuclide concentrations caused by different crop irrigation levels. It is reasonable to use an average annual irrigation rate to estimate long-term radionuclide concentrations in surface soil. This concentration is used to evaluate radionuclide uptake from soil by plants, and to evaluate inhalation, inadvertent soil ingestion, and external exposures. Using this simplification, the average activity concentration of a radionuclide in the surface soil does not depend on the crop type.

METHODS

The mathematical representation of a radionuclide addition and removal processes in the surface soil is expressed by the following differential equation and the initial condition (BSC 2004).

$$\begin{cases} \dfrac{dCs_i(t)}{dt} = Cw_i(t)\, IR(t) - (\lambda_{d,i} + \lambda_{l,i} + \lambda_e)\, Cs_i(t) \\ \\ Cs_i(t) = 0, \ when\ t = 0 \end{cases} \qquad \text{(Eq. 1)}$$

where

- $Cs_i(t)$ = activity concentration of radionuclide i in surface soil per unit area at time t (Bq/m^2)
- i = primary radionuclide index, used for entire biosphere model
- t = time variable (yr)
- $Cw_i(t)$ = activity concentration of radionuclide i in the groundwater at time t (Bq/m^3)
- $IR(t)$ = annual average irrigation rate on land (annual irrigation rate) (m/yr)
- $\lambda_{d,i}$ = radioactive decay constant for radionuclide i (/yr); this can be calculated from radionuclide half-life using the conversion $\ln(2)/T_{d,i}$, where $T_{d,i}$ is half-life of radionuclide i (yr)
- $\lambda_{l,i}$ = average annual leaching removal constant for radionuclide i (/yr)
- λ_e = average annual surface soil erosion removal constant (/yr).

When the radionuclide concentration in the groundwater and the crop irrigation rate are not time-dependent, the analytical solution for Equation 1 is expressed as

$$Cs_i(t) = \frac{Cw_i\, IR}{\lambda_{d,i} + \lambda_{l,i} + \lambda_e}\left[1 - e^{-(\lambda_{d,i} + \lambda_{l,i} + \lambda_e)t}\right] \qquad \text{(Eq. 2)}$$

Radionuclides removed from the surface soil to the deep soil by leaching would no longer be available to many of the environmental transport and receptor exposure pathways. The process of leaching contaminants from the surface soil is evaluated using element-specific leaching coefficients. Leaching coefficients are calculated using a relationship developed by Baes and Sharp (1983). The equation for the leaching removal constant, $\lambda_{l,i}$, is expressed as

$$\lambda_{l,i} = \frac{OW}{d \times \theta \left(1 + \frac{\rho}{\theta} Kd_i\right)} \qquad \text{(Eq. 3)}$$

where

OW = crop overwatering rate (m/yr)
d = surface soil depth (m)
θ = volumetric water content of soil (dimensionless)
ρ = soil density (kg/m^3)
Kd_i = solid-liquid partition coefficient for radionuclide i in surface soil (Bq/kg $_{solid}$)/(Bq/m^3 $_{liquid}$) = (m^3 $_{liquid}$ /kg $_{solid}$)

Under natural conditions, the rate of soil removal by erosion generally is in approximate equilibrium with the rate of soil development from soil forming processes, and under these conditions, soil depth is relatively constant (Troeh et al. 1980). Human activities tend to accelerate the rate of soil removal. The removal of surface soil by erosion would result in the loss of radionuclides attached to the soil particles. The rate of radionuclide removal from surface soils is quantified using a surface soil erosion removal constant (λ_e), which is evaluated as

$$\lambda_e = \frac{ER}{d \times \rho} \qquad \text{(Eq. 4)}$$

where

ER = annual average erosion rate for the surface soil (kg/(m^2 yr))

The term $\lambda_{d,i} + \lambda_{l,i} + \lambda_e$ in Equation 2 can be replaced with one parameter, the effective removal constant, $\lambda_{eff,i}$. Thus, Equation 2 can be expressed as

$$Cs_i(t) = \frac{Cw_i \, IR}{\lambda_{eff,i}} \left(1 - e^{-\lambda_{eff,i} t}\right) \qquad \text{(Eq. 5)}$$

The effective removal constant is an important parameter that determines the rate at which radionuclides approached equilibrium concentrations in the surface soil. Radionuclide concentrations eventually would reach the equilibrium concentration, Cs_i, which is expressed as the ratio of the radionuclide addition rate to the effective removal constant

$$Cs_i = \frac{Cw_i \, IR}{\lambda_{eff,i}} \qquad \text{(Eq. 6)}$$

The radionuclide concentration in surface soil calculated from Equations 2, 5 and 6 are given in units of activity per unit area (Bq/m^2), but it can be converted to activity concentration in Bq per unit mass or volume using soil density and depth of surface soil. The radionuclide concentration in soil is an important parameter for dose assessment, because it contributes all exposure pathways by dust resuspension for inhalation, crop root uptake for ingestion, and ground shine

for external exposure. The equilibrium method is an easy and conservative approach. It is adopted by many long-term radiation dose assessment, such as BIOMASS program (BIOMASS 2000) and Yucca Mountain project (Wu et al. 2003).

This method assumes that irrigation rate and radionuclide concentration in water is a constant at any time for a period of time prior to the irrigation. This period of time is radionuclide dependent, ranging from a few years for technetium to a thousand years for plutonium. To evaluate the conservatism built in the method, a numerical method is used to solve the Equation 1 using a time-dependent radionuclide concentration in water as well as time-dependent irrigation rate. Four radionuclides are selected as representative radionuclides for the evaluation. They are technetium-99, neptunium-237, plutonium-239 and amerisium-241. Typical input values used in the evaluation are listed in Table 1 (BSC 2004).

Table 1. Input parameters and their calculated equilibrium concentration in soil

Parameter	Units	Tc-99	Np-237	Pu-239	Am-241
Kd Value, $K_{d\,i}$	L/kg	0.14	25	1200	2000
Irrigation Rate, $IR(t)$	m/yr	1			
Water Concentration, $Cw_i(t)$	Bq/m^3	1			
Surface Erosion, ER	$kg/m^2/yr$	0.49			
Soil Density, ρ	kg/m^3	1500			
Soil Depth, d	m	0.15			
Over-watering Rate, OW	m/yr	0.079			
Half-life, $T_{d,i}$	yr	2.13E+05	2.14E+06	24100	433
Water content, θ	-	0.23			
Decay Constant, $\lambda_{d,\,i}$	1/yr	3.25E-06	3.24E-07	2.88E-05	1.60E-03
Erosion Constant, λ_e	1/yr	2.18E-03			
Leaching Constant, $\lambda_{l,\,i}$	1/yr	1.20E+00	1.40E-02	2.93E-04	1.76E-04
Effective Removal, $\lambda_{eff,i}$	1/yr	1.20E+00	1.61E-02	2.50E-03	3.95E-03
Equilibrium Soil Concentration, $Cs_i(t)$	Bq/m^2	0.83	62.0	400	253

A numerical model was built in GoldSim software (GoldSim Technology Group, 2003), as shown in Figure 1. Verification was performed using a special case (Case 0) with constant irrigation rate (1 m/yr) and constant radionuclide concentration in water (1 Bq/m^3), whose results can be calculated using Equation 5. The comparison indicates that the results from the numerical method agree with those from the analytical method. Besides the constant irrigation and radionuclide concentration case, other five cases were created based on Case 0 to simulate various situations described as follows.

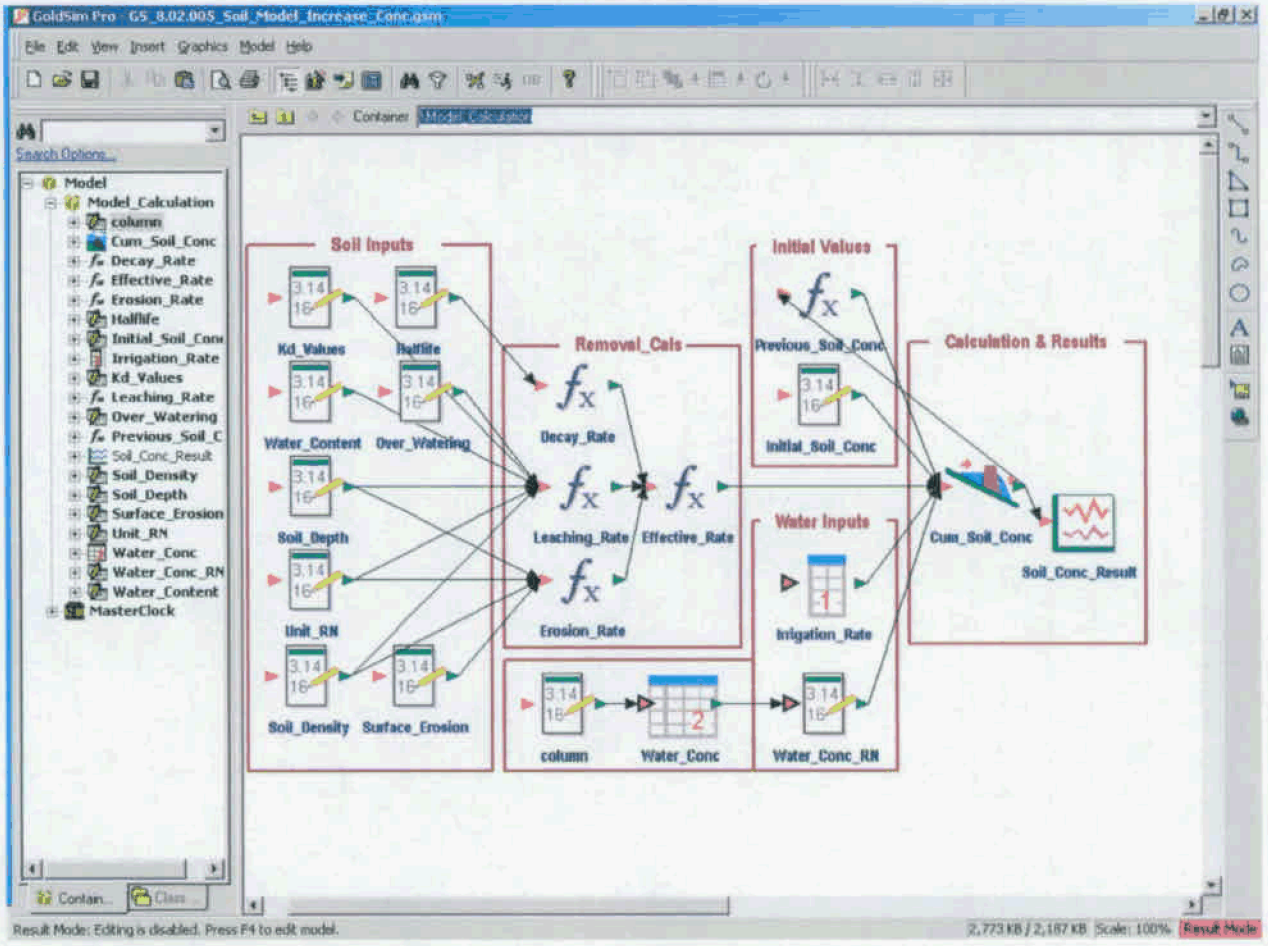

Figure 1. The numerical model built in GoldSim software

The first case is to use radionuclide concentration in water linearly increasing with time from 0 Bq/m^3 at time 0 yr to 1 Bq/m^3 at 10,000 yr. The radionuclide concentration in water is the same as that in Case 0 at 10,000 year. This simulates a simplified case that radionuclides release from the repository slowly increases with time. The second case is to use irrigation rate linearly decreasing with time from 1 m/yr at time 0 yr to 0.5 m/yr at 10,000 yr. The irrigation rate is the same as that in Case 0 at 0 year, but only half of that in Case 0 at 10,000 year. This case simulates a climate change condition where irrigation is reduced to response a cooler and wetter climate in future. The third case is to use radionuclide concentration in water linearly increasing with time and irrigation rate linearly decreasing with time. This case combines Case 1 and Case 2 above. The fourth case is to use irrigation on and off every fifty years with its rate of 1 m/yr. Although irrigation rate is the same as that in Case 0, irrigation occurs only a half of the period. This case simulates the land use condition, because an assumption that a land is in use for the entire period of time is not realistic. The fifth case is to use a model output for the radionuclide concentration in water. However, each radionuclide concentration in water was normalized to its highest point in the period as 1 Bq/m^3 in order to compare with the above-discussed cases (see Figure 2).

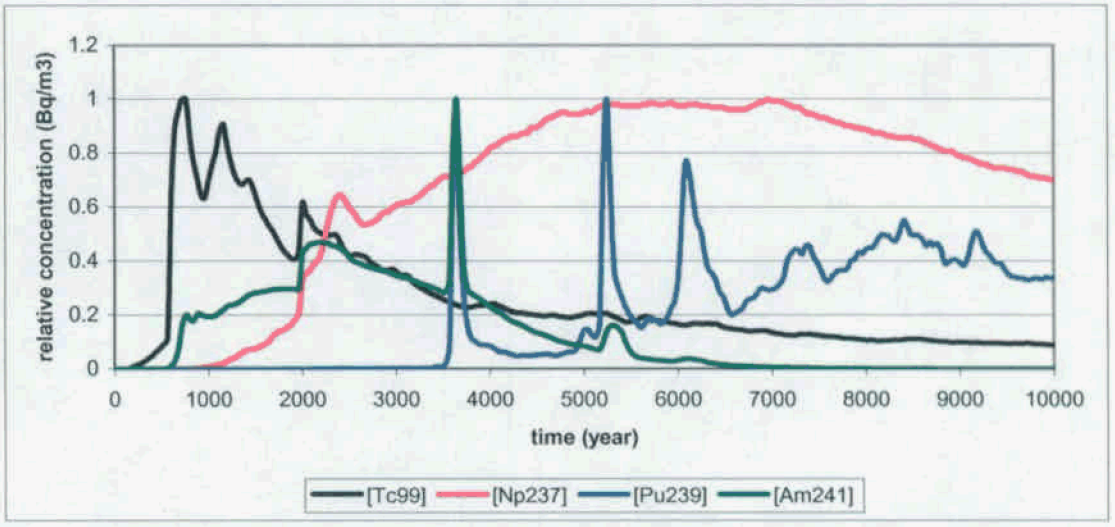

Figure 2. Relative radionuclide concentration in water used in Case 5

RESULTS AND DISCUSSIONS

As mentioned previously, four radionuclides were selected as representative radionuclides. Technetium-99 is soluble in water and has a low K_d value; its effective removal constant is high. It will reach equilibrium in soil in a few years. Neptunium-237 is also soluble in water with relatively low K_d value. It will reach equilibrium in soil in a hundred years. Plutonium-239 has a high K_d value, which causes a low leaching. It will accumulate in soil in a thousand years. Amerisium-241 has a high K_d value, but its half-life is relatively short. It will accumulate in soil in a few hundred years. In addition, these four radionuclides are most important radiation dose contributors in the dose assessment. The simulation results of the six cases are shown in Figure 3 for Tc-99, Figure 4 for Np-237, Figure 5 for Pu-239, and Figure 6 for Am-241.

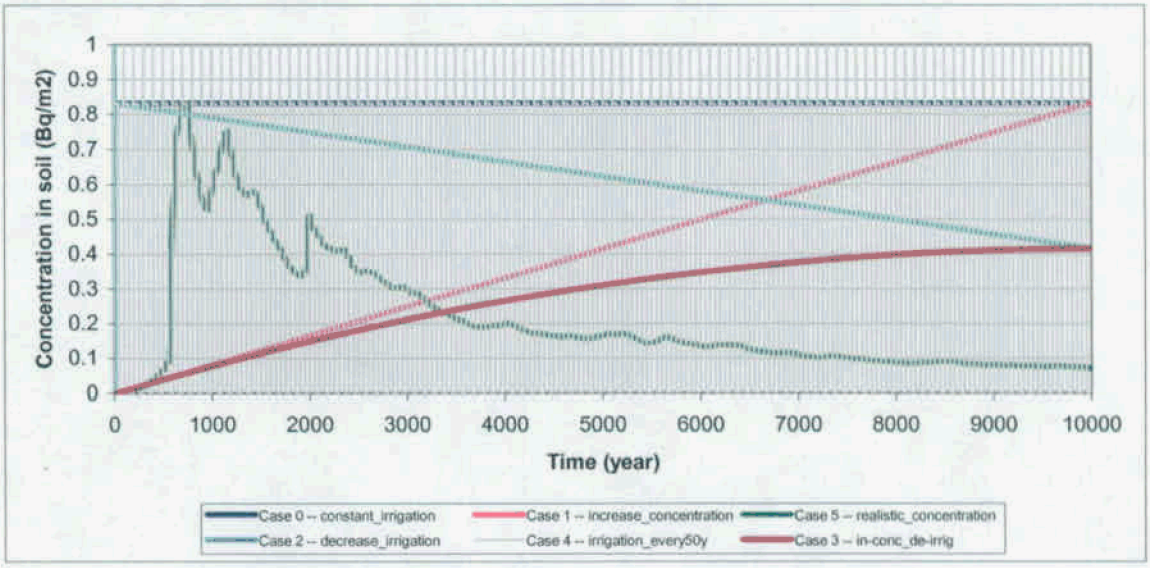

Figure 3. Tc-99 concentration in soil for the six cases

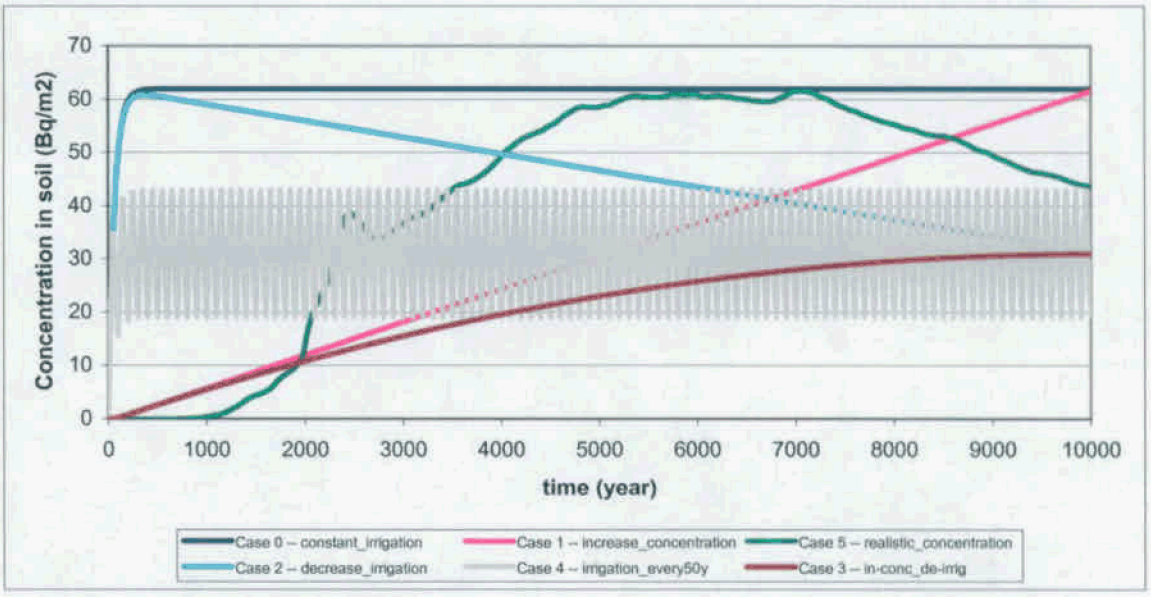

Figure 4. Np-237 concentration in soil for the six cases

For case of radionuclide concentration in water linearly increasing with time (Case 1), radionuclide concentrations in soil also increase with time. At the end of 10,000 years, radionuclide concentrations in soil are very close to those calculated with a constant radionuclide concentration in water (Case 0). For case of irrigation rate linearly decreasing with time, radionuclide concentrations in soil also decrease with time (Case 2). The highest radionuclide concentrations in soil reach at the earlier time. At the end of 10,000 years, radionuclide concentrations in soil are about a half of those calculated with a constant irrigation rate (Case 0). Once again, for a combined case (Case 3), radionuclide concentrations in soil slowly increase to reach the results from Case 2, as expected.

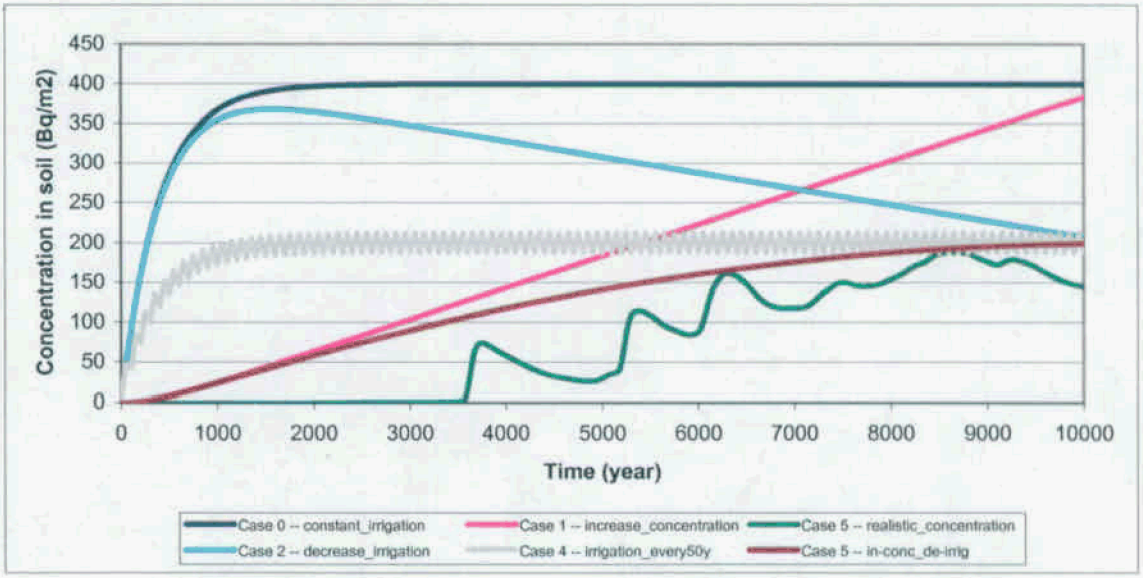

Figure 5. Pu-239 concentration in soil for the six cases

This indicates that a slow change of radionuclide concentration in water or irrigation rate will cause similar change of radionuclide concentration in soil. There is little conservatism by simply using an equilibrium condition, that is, the equilibrium condition in soil will be reached if change of radionuclide concentration in water or irrigation rate is relatively slow with time.

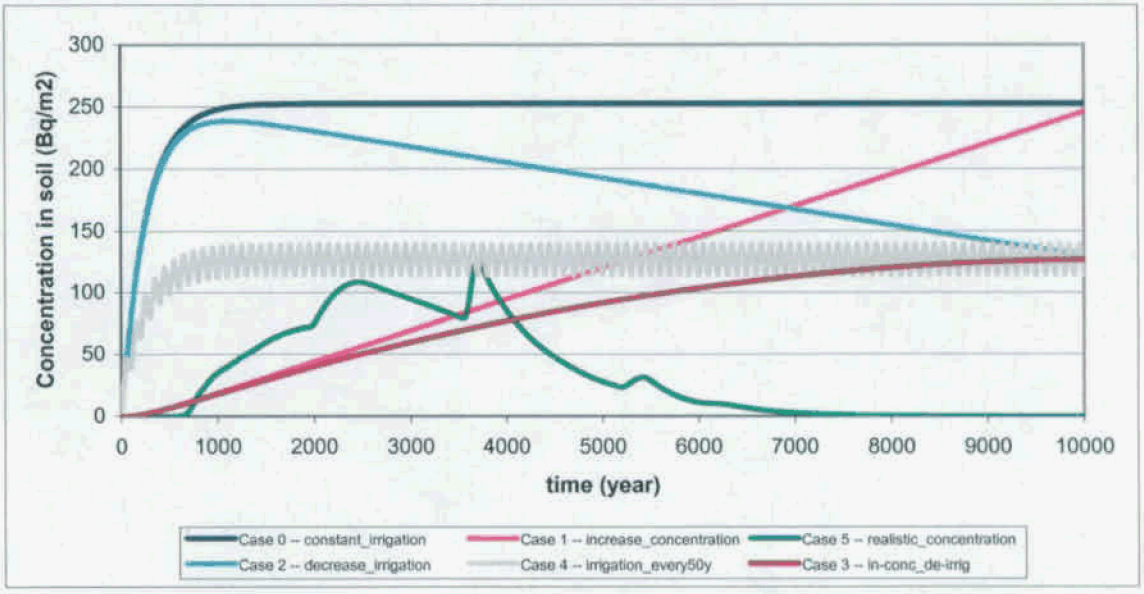

Figure 6. Am-241 concentration in soil for the six cases

For case of irrigation on and off every 50 years (Case 4), radionuclide concentrations in soil strongly depend on radionuclides. For Tc-99, its concentration in soil varies with irrigation pattern from fully equilibrated concentration during irrigation to zero concentration when irrigation is off. For Np-237, concentration in soil oscillates in a relatively large range. However, a small oscillation can be observed for Pu-239 and Am-241 concentrations in soil. The balanced level is about a half of those calculated with a constant irrigation rate (Case 0), which is due to only half of period of time that irrigation occurs.

A very interesting result comes from a realistic radionuclide concentration in water with a constant irrigation rate (1 mg/yr) (Case 5). When an equilibrium condition is assumed, a radionuclide concentration in water would have the same function as a radionuclide concentration in water shown in Figure 2. As can be seen from Figure 3 to Figure 6, soil concentrations of Tc-99 in Figure 3 and Np-237 in Figure 4 have very similar shapes to their water concentrations shown in Figure 2. This is because Tc-99 can reach equilibrium a few year, and Np-237 concentration in water changes slowly. Both radionuclide concentrations in soil can reach the equilibrium concentrations calculated with a constant radionuclide concentration in water ($1 Bq/m^3$) (Case 0).

However, the curves of Pu-239 and Am-241 concentrations in soil (Figures 5 and 6) are very different from corresponding curves in Figure 2. The peaks in Figures 5 and 6 are much flatter than those in Figure 2, due to long-term accumulation in soil. In addition, the highest soil

concentrations of Pu-239 and Am-241 are much lower than those calculated with a constant radionuclide concentration in water (Case 0). Therefore, radionuclide dose at the time of peak concentration in water calculated in the current dose assessment model would be overestimated, because, the highest concentrations in soil are only about a half of those calculated with a constant radionuclide concentration in water (1Bq/m^3) (Case 0).

CONCLUSIONS

It can be concluded that the equilibrium condition in soil will be reached if change of radionuclide concentration in water or irrigation rate is relatively slow with time. In this case, current equilibrium method contains almost no conservatism. However, if a sharp peak occurs in water concentration for a radionuclide that requires long time to accumulate, the calculated dose for that radionuclide could be overestimated using the current equilibrium method.

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